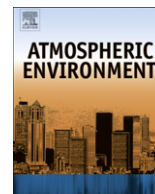


Contents lists available at [SciVerse ScienceDirect](http://www.sciencedirect.com)

Atmospheric Environment

journal homepage: www.elsevier.com/locate/atmosenv

Origin, transport, and vertical distribution of atmospheric pollutants over the northern South China Sea during the 7-SEAS/Dongsha Experiment

Sheng-Hsiang Wang^{a,b,*}, Si-Chee Tsay^b, Neng-Huei Lin^a, Shuenn-Chin Chang^c, Can Li^{b,d}, Ellsworth J. Welton^b, Brent N. Holben^b, N. Christina Hsu^b, William K.M. Lau^b, Simone Lolli^{b,e}, Chun-Chiang Kuo^f, Hao-Ping Chia^f, Chia-Yang Chiu^a, Chia-Ching Lin^a, Shaun W. Bell^{b,g}, Qiang Ji^{b,d}, Richard A. Hansell^{b,d}, Guey-Rong Sheu^a, Kai-Hsien Chi^h, Chi-Ming Peng^{a,f}

^a Department of Atmospheric Sciences, National Central University, Chung-Li 32001, Taiwan

^b Goddard Space Flight Center, NASA, Greenbelt, MD, USA

^c Taiwan EPA, Taipei, Taiwan

^d Earth System Science Interdisciplinary Center, University of Maryland, College Park, MD, USA

^e JCET, University of Maryland Baltimore County, MD, USA

^f WeatherRisk Inc., Taipei, Taiwan

^g Science Systems and Applications Inc., Lanham, MD, USA

^h Institute of Environmental and Occupational Health Sciences, National Yang Ming University, Taipei, Taiwan

H I G H L I G H T S

- First detailed aerosol optical properties in northern South China Sea were investigated.
- Back-trajectory analysis suggested 52% continental origin and 48% oceanic origin.
- Continental origin, mainly from Chinese coastal areas, with higher aerosol loading.
- Comparison with inland measurements, aerosol evolution and mixture were addressed.
- Upper-level (3–4 km) transport of biomass-burning aerosol was investigated by lidar.

A R T I C L E I N F O

Article history:

Received 20 March 2012

Received in revised form

31 October 2012

Accepted 5 November 2012

Keywords:

Aerosols

Optical properties

Back trajectory

UV lidar

Biomass burning

A B S T R A C T

During the spring of 2010, comprehensive *in situ* measurements were made for the first time on a small atoll (Dongsha Island) in the northern South China Sea (SCS), a key region of the 7-SEAS (the Seven South East Asian Studies) program. This paper focuses on characterizing the source origins, transport processes, and vertical distributions of the Asian continental outflows over the region, using measurements including mass concentration, optical properties, hygroscopicity, and vertical distribution of the aerosol particles, as well as the trace gas composition. Cluster analysis of backward trajectories classified 52% of the air masses arriving at ground level of Dongsha Island as having a continental origin, mainly from northern China to the northern SCS, passing the coastal area and being confined in the marine boundary layer (0–0.5 km). Compared to aerosols of oceanic origin, the fine mode continental aerosols have a higher concentration, extinction coefficient, and single-scattering albedo at 550 nm (i.e., 19 vs. 14 $\mu\text{g m}^{-3}$ in $\text{PM}_{2.5}$; 77 vs. 59 Mm^{-1} in β_e ; and 0.94 vs. 0.90 in ω , respectively). These aerosols have a higher hygroscopicity (f at 85% RH = 2.1) than those in the upwind inland regions, suggesting that the aerosols transported to the northern SCS were modified by the marine environment. In addition to the near-surface aerosol transport, a significant upper-layer (3–4 km) transport of biomass-burning aerosols was observed. Our results suggest that emissions from both China and Southeast Asia could have a significant impact on the aerosol loading and other aerosol properties over the SCS. Furthermore, the complex vertical distribution of aerosols-coinciding-with-clouds has implications for remote-sensing observations and aerosol–cloud–radiation interactions.

© 2012 Elsevier Ltd. All rights reserved.

* Corresponding author. Department of Atmospheric Sciences, National Central University, Chung-Li 32001, Taiwan. Tel.: +886 3 422 7151x65527.

E-mail addresses: shenghsiang.wang@gmail.com, carlo@cc.ncu.edu.tw (S.-H. Wang).

1. Introduction

The South China Sea (SCS) is a low-latitude marginal sea in Southeast Asia (SEA) and directly situated downwind of Asian continental outflow, originating from Eurasia and maritime continents, and a wide range of atmospheric pollutants (e.g., Lin et al., 2007; Wang et al., 2011). Much attention has been given lately to the ubiquitous presence of springtime aerosol particles over the region that can alter radiation budget (e.g., Hsu et al., 2004), disturb regional circulation (e.g., Wang et al., 2007), and possibly affect the summer monsoon onset and intensity (e.g., Lau et al., 2008; Lawrence and Lelieveld, 2010). The understanding of physico-chemical characteristics of atmospheric aerosol particles is critical, especially for studying climate effects (IPCC, 2007). To date, however, knowledge of aerosol particle properties and their spatial and temporal variations, as well as their vertical structure over the SEA, is still relatively coarse, mainly due to significant observability challenges of aerosol particles from *in situ* and remote sensing observations (Reid et al., in press; Campbell et al., in press).

Dongsha Island (about 2 km², 20°42'52" N, 116°43'51" E; hereafter referred to as Dongsha; Fig. 1) is a small atoll in the northern SCS, 400 km southwest off the southern tip of Taiwan and 340 km southeast of Hong Kong. Wang et al. (2011, 2012) show that Asian dust transported to Dongsha is characterized by low level transport and is well-mixed with anthropogenic and maritime aerosols, and may have a significant impact on oceanic biogeochemistry in the northern SCS. The Seven Southeast Asian Studies (7-SEAS) program was designed to facilitate interdisciplinary research on the SEA aerosol environment as a whole, promote international collaboration, and to further develop scientific understanding of the impact of biomass burning on clouds, atmospheric radiation, hydrological cycle, and region climate (Reid et al., in press). In support of the 7-SEAS program, the Dongsha Experiment (<http://aerosol.atm.ncu.edu.tw>) was carried out from 10 March to 19 May 2010 on

Dongsha for characterizing the atmospheric pollutants transported to the northern SCS.

Fig. 1 illustrates the study area and the surrounding major emission source regions. Estimated annual CO emissions (Streets et al., 2003) and fire counts obtained from the MODIS/Terra satellite during March–May 2010 are also shown. Asian anthropogenic emissions are concentrated near coastal areas correlating with human population distribution, while biomass-burning emissions are detected in Indochina. From Dongsha, the opportunity for studying continental outflows, as well as the impact of Indochinese biomass burning was available. Hence, the goal of this paper is to study the origin, transport, and vertical distributions of atmospheric pollutants over the northern SCS and their temporal variation. Brief descriptions of measurements made and methods used in the analysis are given in Section 2. The analyses of meteorology, transport regimes, aerosol optical properties, and their implications for regional climate are examined in Section 3, and finally a summary is given in Section 4.

2. Methodology

2.1. Measurements and data

A suite of instruments was deployed at Dongsha Island during the experiment, including the NASA/Goddard Space Flight Center/COMMIT (Chemical, Optical, and Microphysical Measurements of In-situ Troposphere; <http://smartlabs.gsfc.nasa.gov>) mobile observatory, the Taiwan Environmental Protection Administration mobile facility, and a portable and automatic eye safe lidar system. Table 1 lists the instruments and data examined in this study. The instrumentation and calibration of the COMMIT laboratory have been described in detail elsewhere (Jeong et al., 2008; Li et al., 2010; Li et al., in this issue). All *in situ* data were averaged to 1-h intervals. Note, however, that extremely large surface particle concentrations

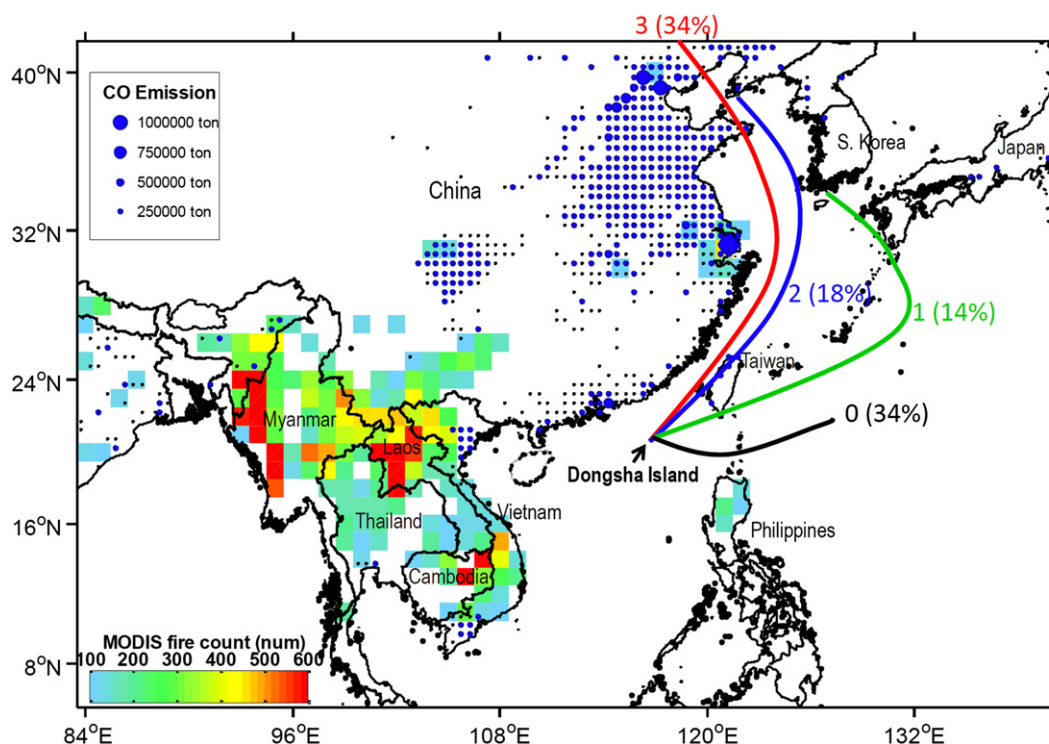


Fig. 1. The location of Dongsha Island in the South China Sea. The blue dot and its size denote the annual CO emission based on Streets et al. (2003). Also shown are the gridded fire counts during March–May 2010 from the MODIS instrument aboard the Terra satellite (<http://disc.sci.gsfc.nasa.gov/neespi/data-holdings/mod14cm1.shtml>). The paths with occurrence percentage of backward trajectories ending at Dongsha are denoted, as described in the text.

Table 1

Summary of measurements and data used in this study.

Measurements	Technique/instrument	Date set
Meteorology	Weather sensors/VAISALA WXT520	Temperature, pressure, RH, wind, precipitation
PM ₁₀ , PM _{2.5}	PM monitor/R&P TEOM 1400a	Mass concentration ^a
CO, O ₃ , SO ₂	Trace gas analyzers/Thermo	Trace gas concentrations
Aerosol optical properties	PSAP (467, 530, and 660 nm) Nephelometer (450, 550, and 700 nm)/TSI Nephelometers (530 nm)/RR	Absorption coefficient ^b Scattering coefficient ^b
Aerosol profiling	EZ-Lidar (355 nm)/Leosphere	Normalized relative backscattering
Aerosol loading	Sun photometer/Cimel	AOD and AE

^a Sample flow was heated to 50 °C.^b Data was reported under standard temperature and pressure (25 °C, 1013.25 hPa).

measured (i.e. the maximum PM₁₀ of 570 µg m⁻³) during an unexpected rare dust episode (1100–2000 UTC March 21, 2010; see Wang et al., 2011) were excluded so that the results discussed here are more representative of climatological conditions.

Optical properties were measured for particles with aerodynamic diameter <2.5 µm. Aerosol scattering coefficients (β_s) at wavelength (λ) = 450, 550, and 700 nm were measured with an integrating nephelometer. Aerosol absorption coefficients (β_a) at λ = 467, 530, 660 nm were measured with a particle soot absorption photometer. Aerosol scattering Ångström exponent (SAE; derived from β_s at λ = 450 and 700 nm) and absorption Ångström exponent (AAE; derived from β_a at λ = 467 and 660 nm), were calculated to interpret particle size and type. β_a at λ = 550 nm were solved through interpolation based on AAE. Aerosol extinction coefficient (β_e ; $\beta_a + \beta_s$) and single-scattering albedo (ω ; β_s/β_e) were then calculated for λ = 550 nm. Statistics of β_s , β_a , β_e , AAE, SAE, and ω are performed using a collocated data set (>87% of available data) of two instruments. Aerosol hygroscopicity was derived based on β_s at λ = 530 nm, measured using two nephelometers with different relative humidity (RH) settings (i.e., ambient and dry RH), where the aerosol hygroscopic factor, $f(\text{RH})$, is then defined as:

$$f(\text{RH}) = \frac{\beta_{s,\text{ambient}}}{\beta_{s,\text{dry}}} \quad (1)$$

An ultraviolet (UV) lidar system, the EZ-Lidar (cf. Lolli et al., 2011), was deployed at Dongsha to profile the optical properties of aerosols and clouds during the experiment. The solid-state pulsed laser source used is the ND:YAG tripled to λ = 355 nm with a repetition rate of 20 Hz. Backscatter signal with a vertical resolution of 15 m was collected. The lidar system includes linear polarization sensitivity. Qualitative information about particle asphericity may be interpreted from the normalized ratio of the perpendicular polarization channel power relative to that of the parallel channel, here referred to as NPPR (e.g., Sassen, 1991). In addition to lidar, an AERONET sun/sky photometer (Holben et al., 1998; <http://aeronet.gsfc.nasa.gov/>) was deployed to measure column-integrated aerosol optical properties, including aerosol optical depth (AOD at 340, 380, 440, 500, 675, 870, 940, and 1020 nm) and Ångström exponent (AE). The AERONET Level-2 AOD at 355 nm was gone through interpolation between 340 and 380 nm.

355 nm AOD was applied for inversion of the lidar equation solving backscatter and extinction coefficients. Under the hypothesis of single scattering, for the case of a vertically-pointing lidar, the detected elastic signal as a function of altitude can be expressed as:

$$P(r) = \frac{C \cdot O(r) \cdot E}{r^2} \cdot [\beta_m(r) + \beta_p(r)] \cdot e^{-2 \int_0^r [\sigma_m(r') + \sigma_p(r')] dr'} \quad (2)$$

where $P(r)$ is the received power from range r ; C denotes the “lidar constant”, which depends on instrumental parameters such as receiver telescope aperture and efficiency of the quantum detectors and the optics; $O(r)$ is the overlap function; E is laser energy. Above 200 m, the field of view of the EZ-Lidar telescope and source intersect and $O(r) = 1$ (cf. Lolli et al., 2011). $\beta_m(r)$ and $\beta_p(r)$ are, respectively, the volume molecular (Rayleigh) and particulate (Mie) backscattering coefficients. $\sigma_m(r)$ and $\sigma_p(r)$ are the molecular and particulate extinction coefficients within distance of r , respectively.

Inverting Eqn. (2) is necessary, because there are two unknowns: the particulate backscattering and the extinction coefficients. Molecular terms may be approximated from balloons and profiles of molecular density in clear air. A common solution (e.g., Fernald, 1984) for Eqn. (2) relies on the assumption of proportionality between these two variables, referred to as the lidar ratio (i.e. extinction-to-backscatter ratio), as well as general turbidity of the mixed aerosol layer. The lidar ratio associated with aerosol types exhibits large variability (e.g., Ackermann, 1998). A mean value of $C = 2.3 \pm 0.2 \times 10^8 \text{ Vm}^2$ was measured for the case study period of 21–24 March 2010 (see Section 3.4). This value was then used to retrieve the aerosol backscattering and extinction profiles, as well as the lidar ratio. A more detailed application of the lidar equation and Fernald retrieval method can be found elsewhere (Welton et al., 2002).

2.2. Trajectory analysis

The Hybrid Single-Particle Lagrangian Integrated Trajectory (HYSPPLIT, Version 4.9) model developed by NOAA Air Resources Laboratory (Draxler and Rolph, 2003) was applied to calculate air mass backward trajectories and help estimate their similarity. The meteorological data used to initialize HYSPPLIT was obtained from the NCEP Global Data Assimilation System (GDAS) with one-degree resolution. Five-day backward trajectories ending at ground level (altitude = 0 km) of Dongsha at 00UTC daily for the period of 10 March–19 May 2010 were calculated and subjected to cluster analysis (embedded in the HYSPPLIT software) in order to allocate them into groups of similar length, origin and transit.

Four clusters (i.e., Clusters 0–3) were identified in this study. The mean trajectories of each cluster are overlaid in Fig. 1, whereas individual trajectories representing each cluster are shown in Fig. 2. Cluster 3 accounts for 34% of total trajectories and represents the longest path, originating from elevated regions over northern China and Mongolia and passing through the coastal areas of China. A similar trajectory result derived investigation the origin of air masses over the Yangtze Delta region of China has been reported by Liu et al. (2012). From this speculation, the air quality at Dongsha can be directly related to upwind inland regions. Trajectories in Cluster 2 (18%) travel mainly along the Chinese coast, but over a much shorter distance. Clusters 2 and 3 both reflect air masses having traversed over coastal areas of East Asia, and associated with anti-cyclonic circulation. Cluster 2 consists of trajectory primary confined below 2 km, while cluster 3 exhibits greater variability with respect to origin heights. Air parcels transport over a highly polluted area in low altitude. Entrainment of air pollutants along each trajectory is considered more likely. In contrast, trajectories within Cluster 1 (14%) encountered a vast maritime area in the western Pacific, and are less likely to consist of relatively new pollutant composition, given the proximally away from the populated coastlines. All other trajectories were classified as Cluster 0 (34%), being mostly oceanic

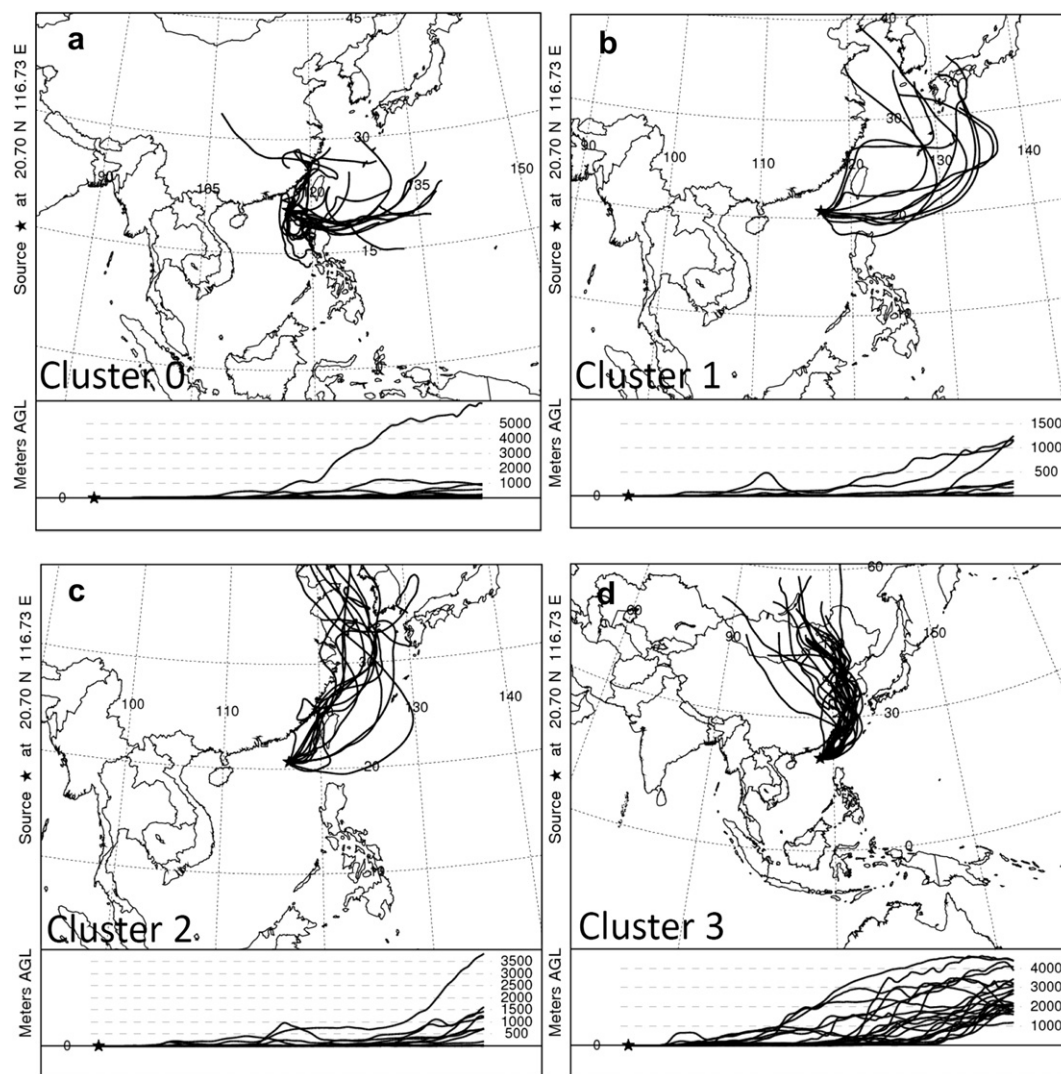


Fig. 2. (a)–(d) HYSPLIT cluster plots for 5-day back trajectories ending at ground level of Dongsha for the period from 10 March to 19 May 2010.

in pathway (e.g., easterly and southerly paths), and originating locally near the northern SCS.

3. Results and discussion

3.1. Meteorology and transport regime

Previous studies (e.g., Yienger et al., 2000; Liu et al., 2003) address that the Siberian anticyclones (the so-called “Siberian Highs”) and mid-latitude cyclones dominate the synoptic weather pattern over East Asia during springtime and play an important role in redistributing Asian emissions downwind. When a mid-latitude cyclone passes East Asia, the associated cold front sweeps through the region to the south, and a Siberian high often follows from the west. Transport behind cold fronts off the Asian continent, in fact, has been considered a major transport mechanism of Asian pollution and dust (e.g., Carmichael et al., 2003; Liang et al., 2004).

Our data show consistent trends for decreasing temperature, increasing pressure and RH associated with the passage of cold front (Fig. 3a). In addition, the varying sequence of trajectory clusters (Fig. 3b) corresponds well with changes in synoptic conditions. For example, when a cold front advances off the Asian continent toward Dongsha, the penetrating low-level

northeasterlies reach the northern SCS, as depicted by the trajectory path of Cluster 2 (e.g., March 26). In the following days (e.g., March 27–29), a southeastward-moving Siberian High approach and the transport pattern reflects of Cluster 3. Finally, after the system moves to the western Pacific, reemerge of Clusters 0 or/and 1 (e.g., March 31–April 2) depending on the location of the high-pressure system. Of course, as spring progressed, this scenario became less frequent, and condition more erratic approaching May and the summer rain season.

3.2. Temporal characteristics of trace gases and aerosols

Temporal variations in trace gas (i.e., CO, O₃, and SO₂) concentrations, PM₁₀ (i.e., particles with size less than 10 μm) mass concentration vs. wind speed at Dongsha are shown in Fig. 4. Over the two-month period, mean values for CO, O₃, SO₂, PM₁₀, and PM_{2.5} were 237.9 ppb, 42.0 ppb, 0.25 ppb, 30.2 μg m⁻³, and 16.6 μg m⁻³, respectively. These mean results, sample medians and standard deviation are summarized in Table 2. The large standard deviations derived indicate that the air quality of this remote site was frequently perturbed by transported air pollutants. Compared to springtime measurements at Cape Hedo (26.8°N, 128.2°E), Japan in the western Pacific for example (e.g.,

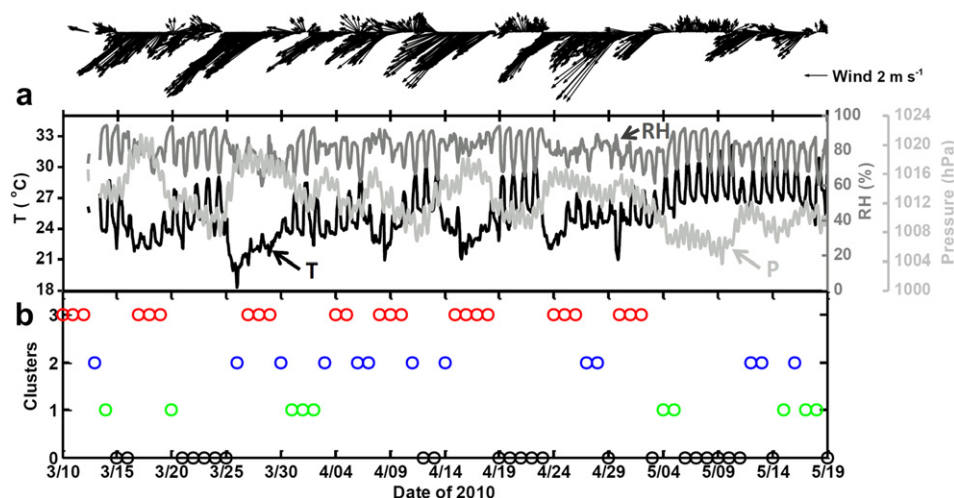


Fig. 3. Time series of (a) wind, temperature, pressure, and relative humidity, and (b) clusters of daily backward trajectories corresponding to Fig. 2 during the Dongsha Island experiment.

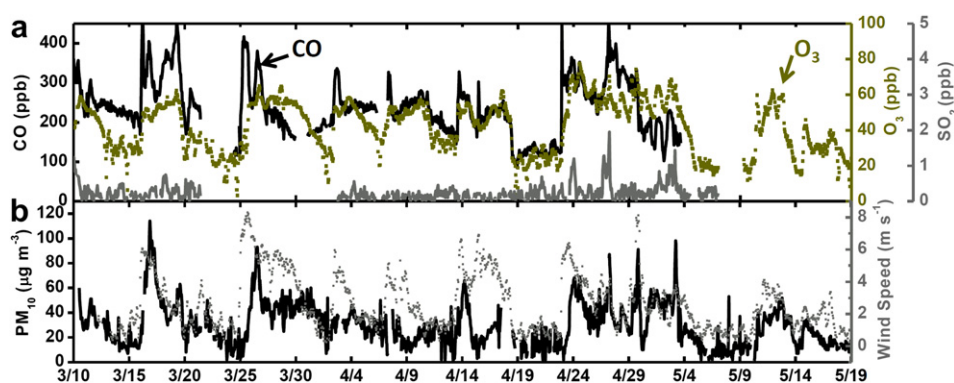


Fig. 4. Time series of (a) CO, O₃, and SO₂ concentration, (b) PM₁₀ mass concentrations and wind speed.

Suthawaree et al., 2008; Sato et al., 2008), CO and O₃ concentrations at Dongsha were comparable, but PM concentrations were lower. Relatively high PM concentrations at Cape Hedo could be attributable to long-range transported Asian dusts. Concentrations of SO₂ were considerably lower than those observed at a rural site in northwestern China (3.4 ppb; Li et al., 2010). Relatively low SO₂ may be caused by efficient oxidation of SO₂ to form sulfuric acid (Chuang et al., in this issue; Bell et al., in this issue), particularly for transport that coincides with clouds (e.g., Tu et al., 2004; Hatakeyama et al., 2011).

Increasing springtime pollutants at Dongsha were more pronounced during transport events from the continent. Fig. 4a and b illustrate that the coincidence of elevated measurements consistently corresponding with the passage of high-pressure systems. In general, concentrations of trace gases and aerosols in Clusters 2 and

3 (hereafter referred to as of continental origin) tend to be higher than in Clusters 0 and 1 (hereafter referred to as of oceanic origin) (Table 3). Note that aerosol particles sampled from Clusters 0 and 1, although oceanic in origin, are more specifically mixtures of sea-salt and other anthropogenic components (e.g., black carbons; Chuang et al., in this issue). Air pollutants of continental origin exhibited higher concentrations (CO: 258.5 ± 60.6 ppb; O₃: 51.2 ± 9.3 ppb; PM₁₀: 37.7 ± 17.1 µg m⁻³; PM_{2.5}: 19.0 ± 8.8 µg m⁻³) than those associated with oceanic origin (CO: 200.0 ± 70.1 ppb; O₃: 31.1 ± 11.9 ppb; PM₁₀: 22.0 ± 14.0 µg m⁻³; PM_{2.5}: 13.6 ± 6.9 µg m⁻³). Spikes in CO and PM₁₀ often appeared right behind a frontal system (i.e. before the approach of a high-pressure system) and were usually associated with Cluster 2 trajectories. Following those spikes, relatively stable high pollutant levels were measured for at least 2–3 days, from northerly flow represented by Cluster 3. Such transport usually lasted until the flow from the ocean (Clusters 0 and 1) displaced the northerly circulation.

The relationship between O₃ and CO concentrations exhibited positive correlations during the Dongsha Experiment, suggesting that the enhancement of O₃ is likely the result from the photochemical O₃ production as described in Suthawaree et al. (2008). Here we further define ΔCO and ΔO₃ to be those concentrations estimated as being contributed by transport only (i.e., the background concentration subtracted from the measured concentration, where background concentrations for CO and O₃ were estimated to be 100 and 15 ppb). Fig. 5 further shows the

Table 2

Statistics of trace gases and aerosol concentrations during March 10–May 19, 2010. N denotes number of hourly data. SD means standard deviation.

Measurements	N	Mean (±SD)	Median	Maximum
CO (ppb)	1185	237.9 (±70.0)	234.0	490.7
O ₃ (ppb)	1590	42.0 (±14.6)	45.3	77.8
SO ₂ (ppb)	947	0.25 (±0.22)	0.19	1.95
PM ₁₀ (µg m ⁻³)	1531	30.2 (±17.5)	27	114.0
PM _{2.5} (µg m ⁻³)	1302	16.6 (±8.5)	16	59.0

Table 3
Mean and standard deviation of trace gases, aerosol concentrations, and aerosol optical properties for Clusters 0–3, oceanic origin (Clusters 0 and 1), and continental origin (Cluster 2 and 3) air masses.

Measurements	Cluster 0	Cluster 1	Cluster 2	Cluster 3	Oceanic	Continental
CO (ppb)	197.2 (± 77.9)	208.1 (± 38.9)	282.6 (± 55.5)	250.4 (± 60.5)	200.0 (± 70.1)	258.5 (± 60.8)
O ₃ (ppb)	29.9 (± 12.8)	33.5 (± 9.2)	46.0 (± 10.7)	53.9 (± 7.0)	31.1 (± 11.9)	51.2 (± 9.3)
PM ₁₀ ($\mu\text{g m}^{-3}$)	19.9 (± 13.8)	26.4 (± 13.4)	34.8 (± 14.3)	39.2 (± 18.2)	22.0 (± 14.0)	37.7 (± 17.1)
PM _{2.5} ($\mu\text{g m}^{-3}$)	13.6 (± 7.3)	14.0 (± 6.1)	19.6 (± 6.9)	18.7 (± 9.7)	13.6 (± 6.9)	19.0 (± 8.8)
β_s at 550 nm (Mm^{-1})	53.6 (± 31.5)	51.9 (± 25.2)	81.5 (± 29.3)	67.6 (± 34.1)	53.1 (± 29.7)	72.4 (± 33.2)
β_a at 550 nm (Mm^{-1})	6.0 (± 3.6)	3.9 (± 2.7)	5.1 (± 2.7)	4.0 (± 2.7)	5.4 (± 3.5)	4.4 (± 2.7)
β_e at 550 nm (Mm^{-1})	59.6 (± 32.8)	55.8 (± 26.0)	86.6 (± 30.5)	71.6 (± 36.0)	58.5 (± 30.9)	76.8 (± 34.9)
ω at 550 nm	0.89 (± 0.07)	0.92 (± 0.06)	0.94 (± 0.03)	0.94 (± 0.03)	0.90 (± 0.07)	0.94 (± 0.03)
SAE _{450–700 nm}	1.44 (± 0.36)	1.19 (± 0.55)	1.46 (± 0.22)	1.28 (± 0.46)	1.36 (± 0.44)	1.34 (± 0.40)
AAE _{467–660 nm}	0.60 (± 0.22)	0.52 (± 0.21)	0.53 (± 0.20)	0.60 (± 0.26)	0.58 (± 0.22)	0.58 (± 0.24)

scatterplot of ΔCO and ΔO_3 . The $\Delta\text{O}_3/\Delta\text{CO}$ ratio (linear regression slope of ΔO_3 – ΔCO correlation) represents the extent of atmospheric oxidation in gaseous form during transport (e.g., Price et al., 2004). A higher $\Delta\text{O}_3/\Delta\text{CO}$ ratio implies a longer distance of transport. Our result shows a higher $\Delta\text{O}_3/\Delta\text{CO}$ ratio of Cluster 3 (0.23) compared to that of Cluster 2 (0.17), which is mainly due to relatively high O₃ concentrations of Cluster 3. We suggest that the higher O₃ concentration of Cluster 3 is associated with a lower titration effect by NO due to strong wind speed. The mean $\Delta\text{O}_3/\Delta\text{CO}$ ratio for continental air masses is found to be 0.21, whereas the $\Delta\text{O}_3/\Delta\text{CO}$ ratio of 0.2–0.5 is characteristic of long-range transpacific episodes to the northwest United States (Price et al., 2004).

3.3. Aerosol particle microphysics: hygroscopicity and optical properties

Anthropogenic aerosols transported near the ocean surface typically experience hygroscopic growth, depending on latitude (Seinfeld, 1986). The mean $f(\text{RH})$ from Eqn. (1) was calculated for each 5% RH interval between 50% and 95%, and fitted to the following equation (Li et al., in this issue):

$$f(\text{RH}) = c \left(1 - \frac{\text{RH}}{100} \right)^{-g} \quad (3)$$

The fitted parameters c and g were 0.89 and 0.45, respectively (Fig. 6). During the Dongsha Experiment, $f(85\%)$ [i.e., $f(\text{RH})$ at 85%

RH] of continental origin ranged from 2.0 to 2.3, with a mean value of 2.1 (according to the Eqn. (3)). An earlier study (Wang et al., 2011) measured $f(85\%)$ at 1.86 for a long-range transported dust event at Dongsha. In comparison, the maritime background $f(85\%)$ measured at Cape Verde (16.73°N, 22.93°W) was about 2.5 (Jeong et al., 2008). Lower $f(85\%)$ of ~ 1.6 for rural environments were observed at Phimai, Thailand (Li et al., in this issue), a site influenced by fresh agricultural fires, as well as at the Southern Great Plains (SGP) site in the US (Jeong et al., 2007). A mean $f(85\%)$ of 2.0 was measured in the densely populated eastern part of China (Xu et al., 2002). Liu et al. (2008) reported that $f(80\%)$ in Ganzhou (23.13°N, 113.25°E) were 2.04, 2.29, and 2.68 for urban aerosols, mixed aerosols, and marine aerosols, respectively. Therefore, the scattering of aerosols farther away from the arid areas is more strongly influenced by atmospheric humidity, because of the greater fraction of soluble materials from both anthropogenic and oceanic sources.

Table 3 lists the mean and standard deviation values for aerosol optical parameters (i.e., β_s , β_a , β_e , AAE, SAE, and ω) derived for each cluster and the two combined transport regimes (i.e., oceanic and continental origins). Mean β_s at 550 nm for continental particles was $72.4 \pm 33.2 \text{ Mm}^{-1}$ (10^{-6} m^{-1}), which is about 40% greater than that of the oceanic origin ($53.1 \pm 29.7 \text{ Mm}^{-1}$) but substantially lower than the value ($\sim 202 \text{ Mm}^{-1}$) measured in the source area of Clusters 2 and 3 (i.e. in the Yangtze delta region of China; Xu et al., 2002).

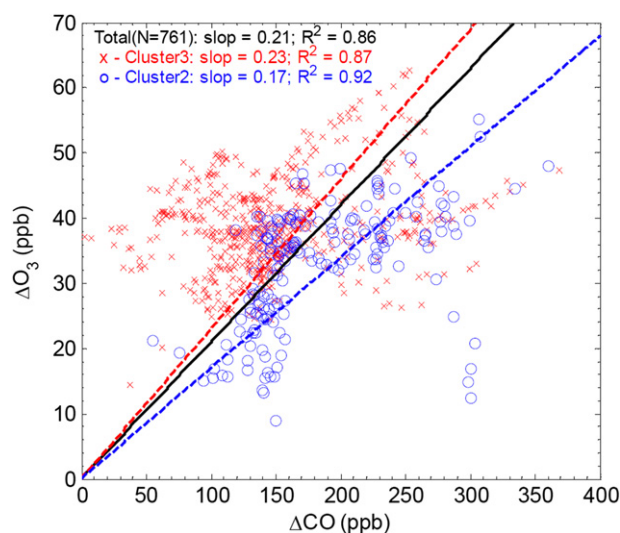


Fig. 5. The scatterplots of ΔCO and ΔO_3 concentrations. The ΔCO and ΔO_3 were defined to the background concentration subtracted from the measured concentration.

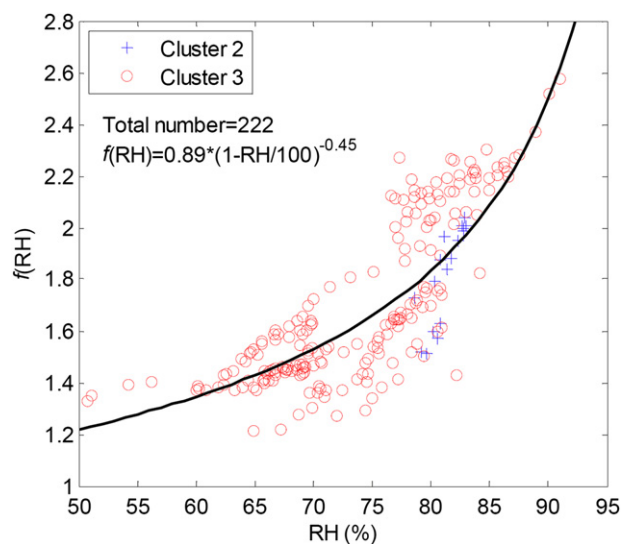


Fig. 6. Aerosol growth factor as a function of relative humidity. The black curve shows the fitting curve suggested in this study.

Mean β_a at 550 nm for aerosols from continental and oceanic origins are 4.4 ± 2.7 and 5.4 ± 3.5 Mm^{-1} , respectively, or about one-twentieth of the value measured at Guangzhou, China (91 Mm^{-1} , Andreae et al., 2008). This in turn results in a lower single-scattering albedo (ω ; 0.9) for aerosols from oceanic origin compared to those from a continental origin (0.94), suggesting moderately absorbing aerosols in both cases. Close examination of those values in each cluster shows a relatively high β_a (6.0 Mm^{-1}) and low ω (0.89 ± 0.07) in Cluster 0 resulting in a stronger absorption characteristic of the oceanic air mass. The trajectories of Cluster 0 are characterized by easterly and southerly paths, and having traversed a shorter distance over maritime continent. Analyses of $\text{PM}_{2.5}$ aerosol components at Dongsha (Chuang et al., in this issue) show that the air mass associated with maritime continent has higher elemental carbon fraction than that associated with continental origin. Nevertheless, strongly absorbing aerosols [i.e., $\omega = 0.83$ (Andreae et al., 2008)] in the vicinity of the northern SCS probably contributed to the background characteristics of the East Asian marginal sea.

Fine mode SAE and AAE in the visible range measured at Dongsha were 1.35 and 0.58, respectively. From comparisons of Clusters 2 and 3, the lower SAE and the higher AAE in Cluster 3 suggest a relatively higher fraction of coarse particles, which is in good agreement with the previous $\text{PM}_{2.5}/\text{PM}_{10}$ ratio described above. It should be noted that although the differences in aerosol optical properties between clusters could reasonably be considered for interpolating aerosol type, as well as source regions, their large standard deviations could bias the conclusions for such a limited sample of data. To better constrain this uncertainty, multi-year measurements are necessary for the future, in order to compile data sets that reflect strongly on document seasonal advection pathway.

3.4. Upper-level transport of Indochinese biomass-burning aerosols

In the prior discussion, we have investigated aerosol composition and particle optical properties measured from Dongsha at the surface and correlated variability with changes in trajectory and upwind origin. At upper levels, however, flow is more westerly. In some situations, biomass-burning particles were observed originating from seasonal burning in Indochina. Utilizing ground-based remote sensing, we show here the preliminary results of the vertical distributions of aerosol particles over Dongsha, as well as their implications on regional climate. Fig. 7 shows EZ-lidar based profiling of particle distributions (i.e., backscattering ratio and NPPR), AOD at $\lambda = 355$ nm, and lidar ratio vs. AE from 21 to 24 March. An attenuated backscatter ratio (i.e. the ratio of the total backscattering coefficient over molecular backscattering coefficient; SR) is used to distinguish the presence of upper-level particle presence.

Single- and two-layer structures are depicted in Fig. 7a and b. Over the 4-day period, near-surface aerosols with high SR were mainly restricted within the marine atmospheric boundary layer (MABL). The MABL top height, which is determined based on the backscattered range corrected signal derivative profile (Flamant et al., 1997), was solved around 0.5 km. In addition to the near-surface aerosol transport, a distinct aerosol layer was clearly visible in the free troposphere ($\sim 3\text{--}4$ km) during 23–24 March. The aerosols in the upper-layer exhibited a higher depolarization, suggesting a different aerosol type than that in the MABL. Analysis of the spatiotemporal variations of MODIS fire counts and trajectory suggested that the upper-level transport was most likely attributed to biomass-burning activities in Indochina (Fig. 8).

Between 1200 and 1900 UTC on 21 March, NPPR data showed two separate layers with a strong depolarizing feature. This,

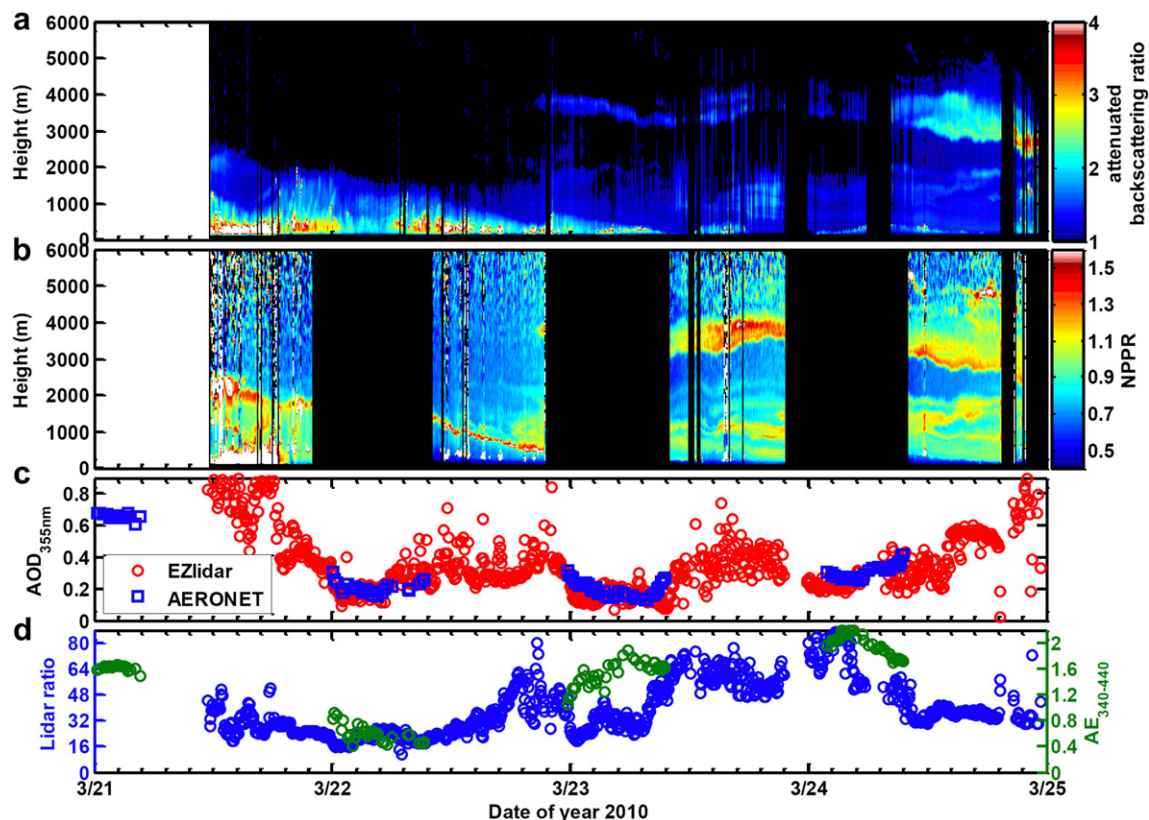


Fig. 7. Time series of vertical distribution of (a) attenuated backscattering ratio, (b) NPPR, (c) AOD at 355 nm, and (d) Lidar ratio vs. $\text{AE}_{340-440 \text{ nm}}$ during 21–24 March 2010. Daytime NPPR measurements are not shown because of signal saturation. LR and AOD values are retrieved profile-by-profile with a temporal resolution of 60 s and averaged every 10 min.

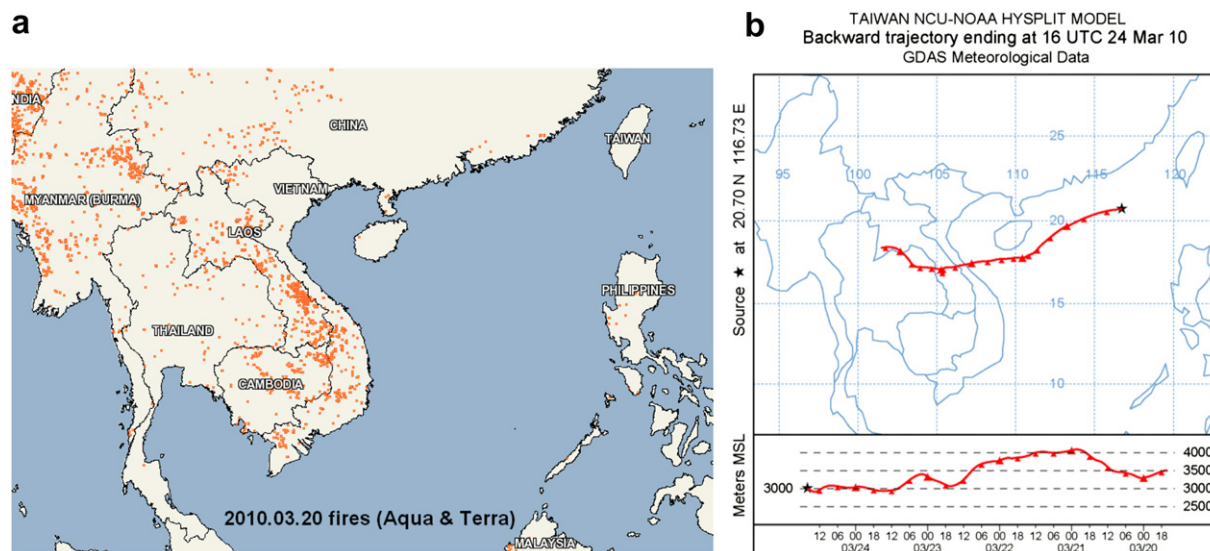


Fig. 8. (a) MODIS fire counts on 20 March 2010 (<http://firms.modaps.eosdis.nasa.gov/firemap/>). (b) HYSPLIT 5-day back trajectories ending at 16 UTC 24 March 2010 at 3000 m for Dongsha.

together with a value of a lidar ratio of 39 ± 4 sr and an AOD > 0.8 , as shown in Fig. 7c and d respectively, was related to a dust episode reported by Wang et al. (2011). On the following day (22 March), lower values of AOD (~ 0.2), lidar ratio (~ 20 sr), and $AE_{340-440\text{ nm}}$ (~ 0.7) were more characteristic of marine aerosols (e.g., Tesche et al., 2011). From late 22 March, with the presence of upper-level aerosol, columnar aerosol properties showed large variation and higher values compared to the maritime background. The measured values of the lidar ratio (up to 74 ± 7 sr) and $AE_{340-440\text{ nm}}$ (up to 2) were in agreement with observations at Cape Verde Island (almost at same latitude of Dongsha) for transported biomass-burning plume (Tesche et al., 2011), and characteristic of absorbing particle type (Ackermann, 1998).

Upper-level transport of biomass-burning aerosols to central Japan and northern Taiwan has been addressed in previous studies (e.g., Murayama et al., 2004; Wang et al., 2010). Here we provide the first ground-based observational evidence that the northern SCS can be an important transport pathway for these smoke plumes. In particular, the presence of advection continental air masses serves to bring relatively stable air from which to decouple advecting smoke layers being driven to the north from southwesterly flow. The wide spread occurrence of multi-layer aerosol transports over East Asia, the northern SCS, and the western Pacific may have important implications for aerosol–clouds–radiation interactions and impact on regional climate. Although Wang et al. (2010) suggest that the clear-sky direct radiative effect is only weakly sensitive to the vertical distribution of aerosols over this region, the coexistence of clouds could significantly amplify the impact of overlying aerosols on the radiation budget. For example, based on satellite observations Hsu et al. (2004) show that the presence of smoke-laden clouds over southern China can reduce the shortwave radiation flux at the top of the atmosphere by 100 W m^{-2} . Thus, earlier investigations of the vertical distribution and optical properties of aerosol provide a knowledge base for further investigating the impact of aerosol over the SCS and surrounding areas.

4. Concluding remarks

In support of the 7-SEAS (the Seven South East Asian Studies) program, the 7-SEAS/Dongsha Island experiment was carried out from 10 March to 19 May 2010 during the Indochina biomass-

burning season. The project sought to investigate, in detail, the characteristics of trace gases and aerosols, and how they relate to their effects on environment variability and regional climate. We analyze data from *in situ* and lidar measurements, and trajectory simulations, to explore the relationships between the origin, transport, and vertical distribution of atmospheric pollutants over the northern South China Sea (SCS).

Mean concentrations of CO, O₃, and PM₁₀ were 238 ppb, 42 ppb, and $30\text{ }\mu\text{g m}^{-3}$, respectively. These measurements are comparable with springtime measurements at Cape Hedo, a remote observatory on Okinawa Island in the western Pacific (e.g., Suthawaree et al., 2008; Sato et al., 2008). However, Dongsha Island is situated at a lower latitude and relatively close to the Asian continent. Analyses of meteorology and back trajectories ending at the surface at Dongsha show that springtime air masses consistently traverse the coastal areas of East Asia, and are strongly related to the northwesterly winds following frontal passages. We are able to correlate many of these events with distinct pollution events at the site. The distinct character of transport regime change is simplified to represent in either maritime background aerosols or increased atmospheric pollutants having a continental origin. Dongsha Island is believed to represent an ideal location for monitoring the southward transport of Asian pollutants offshore.

Fine mode particle (PM_{2.5}) concentrations, scattering and absorption coefficients (at 550 nm), scattering Ångström exponent (SAE), and single-scattering albedo (ω), averaged for those air masses with continental origin, were $19.0 \pm 8.8\text{ }\mu\text{g m}^{-3}$, $72.4 \pm 33.2\text{ Mm}^{-1}$, $4.4 \pm 2.7\text{ Mm}^{-1}$, 1.34 ± 0.40 , and 0.94 ± 0.03 , respectively. Compared to the inland measurements upwind of the northern SCS, aerosol particles observed at Dongsha exhibited lower concentrations, scattering and absorption coefficients, yet they also showed a larger hygroscopicity ($f(85\%) = 2.1$). Results from aerosol optical measurements suggest air masses that were originally contaminated by anthropogenic pollutants, and which were diluted and modified by uptake of marine aerosols while passing over open waters before reaching the site. Although the relationship between atmospheric dynamics and transport is relatively clear, the microphysics and chemical transformation of air pollutants during a long-range transport can be more complicated. We suggest that in order to fully understand the characteristics of atmospheric pollutants over the SCS region, it will be

critical to conduct collaborative multi-annual and regional *in situ* measurements in the future.

Lidar observations indicate that the aforementioned aerosols having a continental origin were mostly confined near and within the marine boundary layer, whereas biomass-burning aerosols from Indochina, upon encountering the colder and dryer continental air, were lifted and sustained within the free troposphere (3–4 km). Changes in columnar aerosol optical properties (i.e., AOD, AE, and lidar ratio) were found in the scenario when the upper-level transport of biomass-burning aerosols overlaid the low-level aerosol layer. The two-layer structure found in this experiment, in terms of aerosol types and optical properties, has important implications for remote-sensing observations and atmospheric radiative transfer. Overall, our results from these island measurements provide essential knowledge on source origins, transport, evolution, and vertical distribution of Asian continental outflows over a low-latitude marginal sea, which serves as a critical element in evaluating the success of the 7-SEAS project.

Acknowledgments

This work was supported by the National Science Council of Taiwan under grants No. 99-2111-M-008-011, 100-2111-M-008-011, 101-2111-M-008-003, and by the Taiwan Environmental Protection Administration under contracts No. EPA-99-FA11-03-A097, EPA-100-U11-02-101 and also by the NASA Radiation Sciences Program managed by Dr. Hal Maring. The authors thank two anonymous reviewers for their useful comments and suggestions.

References

- Ackermann, J., 1998. The extinction-to-backscatter ratio of tropospheric aerosol: a numerical study. *Journal of Atmospheric and Oceanic Technology* 15, 1043–1050.
- Andreea, M.O., Schmid, O., Yang, H., Chand, D., Zhen Yu, J., Zeng, L.-M., Zhang, Y.-H., 2008. Optical properties and chemical composition of the atmospheric aerosol in urban Guangzhou, China. *Atmospheric Environment* 42, 6335–6350.
- Bell, S.W., et al. Constraining aerosol optical models using ground-based, collocated particle size and mass measurements in variable air mass regimes during the 7-SEAS/Dongsha Experiment. *Atmospheric Environment*, in this issue.
- Campbell, J.R., Reid, J.S., Westphal, D.L., Zhang, J., Tackett, J.L., Chew, B.N., Welton, E.J., Shimizu, A., Sugimoto, N., Aoki, K., Winker, D.M. Characterizing the vertical profile of aerosol particle extinction and linear depolarization over Southeast Asia and the Maritime Continent: the 2007–2009 view from CALIOP. *Atmospheric Research*, in press.
- Carmichael, G.R., et al., 2003. Regional-scale chemical transport modeling in support of the analysis of observations obtained during the TRACE-P experiment. *Journal of Geophysical Research – Atmospheres* 108 (D21), 8823.
- Chuang, M.-T., Lin, N.-H., Chang, S.-C., Wang, J.-L., Sheu, G.-R., Chang, Y.-J., Lee, C.-T. Aerosol chemical properties and related pollutants measured at Dongsha Island in the northern South China Sea during 7-SEAS/Dongsha Experiment. *Atmospheric Environment*, in this issue.
- Draxler, R.R., Rolph, G.D., 2003. HYSPLIT (HYbrid Single-Particle Lagrangian Integrated Trajectory) Model Access via NOAA ARL READY Website. NOAA Air Resources Laboratory, Silver Spring, MD. <http://www.arl.noaa.gov/ready/hysplit4.html>.
- Fernald, F.G., 1984. Analysis of atmospheric lidar observations: some comments. *Applied Optical* 23, 652–653.
- Flamant, C., Pelon, J., Flamant, P.H., Durand, P., 1997. Lidar determination of the entrainment zone thickness at the top of the unstable marine atmospheric boundary layer. *Boundary-Layer Meteorology* 83, 247–284.
- Hatakeyama, S., et al., 2011. Aerial observation of aerosols transported from East Asia – chemical composition of aerosols and layered structure of an air mass over the East China Sea. *Aerosol and Air Quality Research* 11, 497–507.
- Holben, B.N., et al., 1998. AERONET – a federated instrument network and data archive for aerosol characterization. *Remote Sensing Environment* 66, 1–16.
- Hsu, N.C., Herman, J.R., Tsay, S.C., 2004. Radiative impacts from biomass burning in the presence of clouds during boreal spring in southeast Asia. *Geophysical Research Letters* 30 (5), 1224.
- IPCC, 2007. In: Solomon, S., et al. (Eds.), Intergovernmental Panel on Climate Change (IPCC), Climate Change 2007. Working Group I Report "The Physical Science Basis". Cambridge Univ. Press, New York, p. 996.
- Jeong, M.-J., Li, Z., Andrews, E., Tsay, S.-C., 2007. Effect of aerosol humidification on the column aerosol optical thickness over the Atmospheric Radiation Measurement Southern Great Plains site. *Journal of Geophysical Research – Atmospheres* 112, D10202.
- Jeong, M.J., Tsay, S.C., Ji, Q., Hsu, N.C., Hansell, R.A., Lee, J., 2008. Ground-based measurements of airborne Saharan dust in marine environment during the NAMMA field experiment. *Geophysical Research Letters* 35, L20805.
- Lau, K.M., et al., 2008. The joint aerosol–monsoon experiment: a new challenge for monsoon climate research. *Bulletin of the American Meteorological Society* 89, 369–383.
- Lawrence, M.G., Lelieveld, J., 2010. Atmospheric pollutant outflow from southern Asia: a review. *Atmospheric Chemistry and Physics* 10, 11017–11096.
- Li, C., et al., 2010. Anthropogenic air pollution observed near dust source regions in northwestern China during springtime 2008. *Journal of Geophysical Research – Atmospheres* 115, D00K22.
- Li, C., et al. Characteristics and composition of atmospheric aerosols in Phimai, central Thailand during BASE-ASIA. *Atmospheric Environment*, in this issue.
- Liang, Q., Jaegle, L., Jaffe, D.A., Weiss-Penzias, P., Heckman, A., Snow, J.A., 2004. Long-range transport of Asian pollution to the northeast Pacific: seasonal variations and transport pathways of carbon monoxide. *Journal of Geophysical Research – Atmospheres* 109, D23S07.
- Lin, I.L., Chen, J.-P., Wong, G.T.F., Huang, C.-W., Lien, C.-C., 2007. Aerosol input to the South China Sea: results from the MODerate resolution imaging spectro-radiometer, the quick scatterometer, and the measurements of pollution in the troposphere sensor. *Deep Sea Research Part II: Topical Studies in Oceanography* 54, 1589–1601.
- Liu, H.Y., Jacob, D.J., Bey, I., Yantosca, R.M., Duncan, B.N., Sachse, G.W., 2003. Transport pathways for Asian pollution outflow over the Pacific: interannual and seasonal variations. *Journal of Geophysical Research – Atmospheres* 108 (D20), 8786.
- Liu, J., Zheng, Y., Li, Z., Flynn, C., Cribb, M., 2012. Seasonal variations of aerosol optical properties, vertical distribution and associated radiative effects in the Yangtze Delta region of China. *Journal of Geophysical Research – Atmospheres* 117, D00K38.
- Liu, X.G., Cheng, Y.F., Zhang, Y.H., Jung, J.S., Sugimoto, N., Chang, S.Y., Kim, Y.J., Fan, S.J., Zeng, L.M., 2008. Influences of relative humidity and particle chemical composition on aerosol scattering properties during the 2006 PRD campaign. *Atmospheric Environment* 42, 1525–1536.
- Lolli, S., Sauvage, L., Loac, S., Lardier, M., 2011. EZ Lidar: a new compact autonomous eye-safe scanning aerosol Lidar for extinction measurements and PBL height detection. Validation of the performances against other instruments and intercomparison campaigns. *Óptica Pura y Aplicada* 44 (1), 33–41.
- Murayama, T., Muller, D., Wada, K., Shimizu, A., Sekiguchi, M., Tsukamoto, T., 2004. Characterization of Asian dust and Siberian smoke with multiwavelength Raman lidar over Tokyo, Japan in spring 2003. *Geophysical Research Letters* 31, L23103.
- Price, H.U., Jaffe, D.A., Cooper, O.R., Doskey, P.V., 2004. Photochemistry, ozone production, and dilution during long-range transport episodes from Eurasia to the northwest United States. *Journal of Geophysical Research – Atmospheres* 109, D23S13.
- Reid, J.S., et al. Observing and understanding the Southeast Asian aerosol system by remote sensing: an initial review and analysis for the Seven Southeast Asian Studies (7SEAS) program. *Atmospheric Research*, in press.
- Sassen, K., 1991. The polarization lidar technique for cloud research: a review and current assessment. *Bulletin of the American Meteorological Society* 72, 1848–1866.
- Sato, K., Li, H., Tanaka, Y., Ogawa, S., Iwasaki, Y., Takami, A., Hatakeyama, S., 2008. Long-range transport of particulate polycyclic aromatic hydrocarbons at Cape Hedo remote island site in the East China Sea between 2005 and 2008. *Journal of Atmospheric Chemistry* 61 (3), 243–257.
- Seinfeld, J.H., 1986. *Atmospheric Chemistry and Physics of Air Pollution*. John Wiley and Sons, New York, 768 pp.
- Suthawaree, J., Kato, S., Takami, A., Kadana, H., Toguchi, M., Yogi, K., Hatakeyama, S., Kajii, Y., 2008. Observation of ozone and carbon monoxide at Cape Hedo, Japan: seasonal variation and influence of long-range transport. *Atmospheric Environment* 42, 2971–2981.
- Streets, D.G., et al., 2003. An inventory of gaseous and primary aerosol emissions in Asia in the year 2000. *Journal of Geophysical Research – Atmospheres* 108 (D21), 8809.
- Tesche, M., Gross, S., Ansmann, A., Müller, D., Althausen, D., Freudenthaler, V., Esselborn, M., 2011. Profiling of Saharan dust and biomass-burning smoke with multiwavelength polarization Raman lidar at Cape Verde. *Tellus B* 63, 649–676.
- Tu, F.H., Thornton, D.C., Bandy, A.R., Carmichael, G.R., Tang, Y.H., Thornhill, K.L., Sachse, G.W., Blake, D.R., 2004. Long-range transport of sulfur dioxide in the central Pacific. *Journal of Geophysical Research – Atmospheres* 109, D15S08.
- Wang, S.H., Lin, N.H., Chou, M.D., Woo, J.H., 2007. Estimate of radiative forcing of Asian biomass-burning aerosols during the period of TRACE-P. *Journal of Geophysical Research – Atmospheres* 112, D10222.
- Wang, S.H., Lin, N.H., Chou, M.D., Tsay, S.C., Welton, E.J., Hsu, N.C., Giles, D.M., Liu, G.R., Holben, B.N., 2010. Profiling transboundary aerosols over Taiwan and assessing their radiative effects. *Journal of Geophysical Research – Atmospheres* 115, D00K31.
- Wang, S.H., et al., 2011. First detailed observations of long-range transported dust over the northern South China Sea. *Atmospheric Environment* 45, 4804–4808.

- Wang, S.-H., Hsu, N.C., Tsay, S.-C., Lin, N.-H., Sayer, A.M., Huang, S.-J., Lau, W.K.M., 2012. Can Asian dust trigger phytoplankton blooms in the oligotrophic northern South China Sea? *Geophysical Research Letters* 39, L05811.
- Welton, E.J., Voss, K.J., Quinn, P.K., Flatau, P.J., Markowicz, K., Campbell, J.R., Spinhirne, J.D., Gordon, H.R., Johnson, J.E., 2002. Measurements of aerosol vertical profiles and optical properties during INDOEX 1999 using micropulse lidars. *Journal of Geophysical Research – Atmospheres* 107 (D19), 8019.
- Xu, J., Bergin, M.H., Yu, X., Liu, G., Zhao, J., Carrico, C.M., Baumann, K., 2002. Measurement of aerosol chemical, physical and radiative properties in the Yangtze delta region of China. *Atmospheric Environment* 36, 161–173.
- Yienger, J.J., Galanter, M., Holloway, T.A., Phadnis, M.J., Guttikunda, S.K., Carmichael, G.R., Moxim, W.J., Levy, H., 2000. The episodic nature of air pollution transport from Asia to North America. *Journal of Geophysical Research – Atmospheres* 105, 26,931–26,945.